

### 3.0 RECYCLED URANIUM MASS FLOW

#### 3.1 RECYCLED URANIUM DESCRIPTION

For purposes of the DOE recycled uranium mass balance project, RU has been defined as any uranium that has been irradiated in a reactor and, as a result, contains TRU material (e.g., Pu and Np), fission products (e.g., Tc), and reactor-generated uranium products ( $^{236}\text{U}$ ). The methodology applied in this Y-12 Complex project for identifying the flow of RU materials includes the criteria of (1) the source site, (2) the isotopic constituents, and (3) the wt-% assays of the material. Sites identified as RU source sites are the U.S. government facilities that operated production reactors and/or used chemical separation processes to extract uranium from irradiated fuel. Primary source sites are the Savannah River Site (SRS), the Idaho Chemical Processing Plant (ICPP), and Hanford. The majority of Y-12 Complex transfers with SRS and ICPP have involved RU (although significant quantities of fresh fuel and sweetener<sup>1</sup> were also shipped to Savannah River). Secondary source sites providing RU materials to the Y-12 Complex are the Oak Ridge Gaseous Diffusion Plant (ORGDP) and the Paducah Gaseous Diffusion Plant (PGDP).

Data for Y-12 Complex material transactions with the RU source sites were extracted from Material Balance Reports (MBRs), shipment and receipt registers, historical summary reports, and individual Nuclear Material Transaction Reports. All of these reports were issued by the site Nuclear Material Control and Accountability (NMC&A) organizations and provide official accountability data for all uranium and for other accountable nuclear materials at each site. Under the Y-12 Complex NMC&A program, uranium is an accountable nuclear material; however, RU is not separately accountable.

The various reports reviewed included the name or symbol code of the accountability station where material was shipped or received, the material type, the amount of uranium, and the  $^{235}\text{U}$  assay. It was necessary to review data at the Nuclear Material Transfer Report level (i.e., forms 101 and 741) to more accurately determine the material type and the  $^{235}\text{U}$  assay because summary reports extracted from the same data often camouflaged the details of the material and assay.

While the methodology used in this project for identifying and tracking RU was the best available, it was imperfect, and so some loss of distinction between RU and non-RU material was unavoidable. Physical losses or discards of RU to the burial ground may have occurred which could not be identified and quantified. Loss of distinction also occurred as a result of blending RU with non-RU materials. Other losses of accountability may have occurred as a result of unavailable or ambiguous data.

Due to these limitations, the Project Team cannot claim with certainty that all activity related to the Y-12 Complex RU shipments, receipts, and inventories has been reviewed. However, the team believes this review is suitably comprehensive to have identified essentially all of the RU streams.

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<sup>1</sup> HEU used to blend with recycled uranium fuel feed to increase its enrichment is referred to as “sweetener.”

## 3.2 URANIUM RECEIPTS

Annual receipts of RU from the primary source sites included highly enriched RU from Savannah River and ICPP and slightly depleted RU from Hanford; these receipts are summarized in Table 3.2-1. Receipts of RU ICPP began in 1953, and receipts from Savannah River began in 1955. Some RU was received by the Y-12 Complex after 1989 but was not processed, primarily due to the Y-12 Complex stand down and the shutdown of the Savannah River reactors, which eliminated the need for recycled fuel.

Beginning in 1955, the Savannah River Site sent highly enriched uranyl nitrate (UN) solution to the Y-12 Complex in tanker trucks with a 3,800 – 5,000 gallon capacity. The concentration of the uranyl nitrate solution received from SRS was approximately 5 g  $^{235}\text{U}$ /liter. After evaporation, the material went through purification by solvent extraction, denitration to produce  $\text{UO}_3$ , reduction to  $\text{UO}_2$ , hydrofluorination to  $\text{UF}_4$ , and “bomb” reduction to metal. The metal was cleaned and packaged for shipment back to Savannah River or placed in storage until Savannah River requested the material.

From 1972 to 1989, Savannah River sent ingot material of uranium-aluminum alloy (U-Al) for processing. This material was processed by first combining it with NaOH solution to dissolve the aluminum, which left sodium diuranate solids. The sodium diuranate was then dissolved in nitric acid, producing uranyl nitrate solution, which was purified and converted to metal. Not all of the U-Al material was processed, and some remains in storage at the Y-12 Complex today.

Savannah River also sent dross and furnace sweepings from the U-Al casting process, which were processed by NaOH dissolution to remove aluminum and then by nitric acid dissolution and finally purification and conversion to metal. Not all of this material was processed, and some remains in storage today at the Y-12 Complex. In total, Savannah River sent 125.2 MT of highly enriched RU to the Y-12 Complex. This flow of material between the Y-12 Complex and Savannah River is depicted in Figure 3.2-1.

The Y-12 Complex received 42.6 MT of slightly enriched RU (0.74%  $^{235}\text{U}$ ) from SRS that was transferred to Fernald. This material was received in five shipments which were transferred without repackaging; since this material did not contribute to personnel or environmental exposure, it is not included in this study.

From 1953 until the early-1990s, the ICPP processed spent Navy, research, and experimental reactor fuel to recover and recycle the HEU. The product of ICPP was sent to the Y-12 Complex for processing to metal and subsequent shipment to Savannah River. Initially, the product was UN solution; however, in 1971, a denitrator was installed, and subsequently, the product was uranium trioxide ( $\text{UO}_3$ ). A total of 25.7 MT of highly enriched RU was received at the Y-12 Complex from ICPP.

The Y-12 Complex received 1.5 MT of slightly depleted RU in the form of  $\text{UO}_3$  from Hanford with an assay of 0.65%  $^{235}\text{U}$ . Because the assay is in the range of recovered uranium product of the Hanford reprocessing plant, it was assumed to be RU. Another 142 MTU received from Hanford was initially classified as RU, but this was determined to be unirradiated slugs returned from the Pile Enrichment Experiment and so was not included in this study.

Annual receipts of RU from sites other than Savannah River, ICPP, and Hanford are summarized in Table 3.2-2. Because the assay of the material received from these secondary sites was in the range of 0.59 to 0.69%  $^{235}\text{U}$ , the material was assumed to be

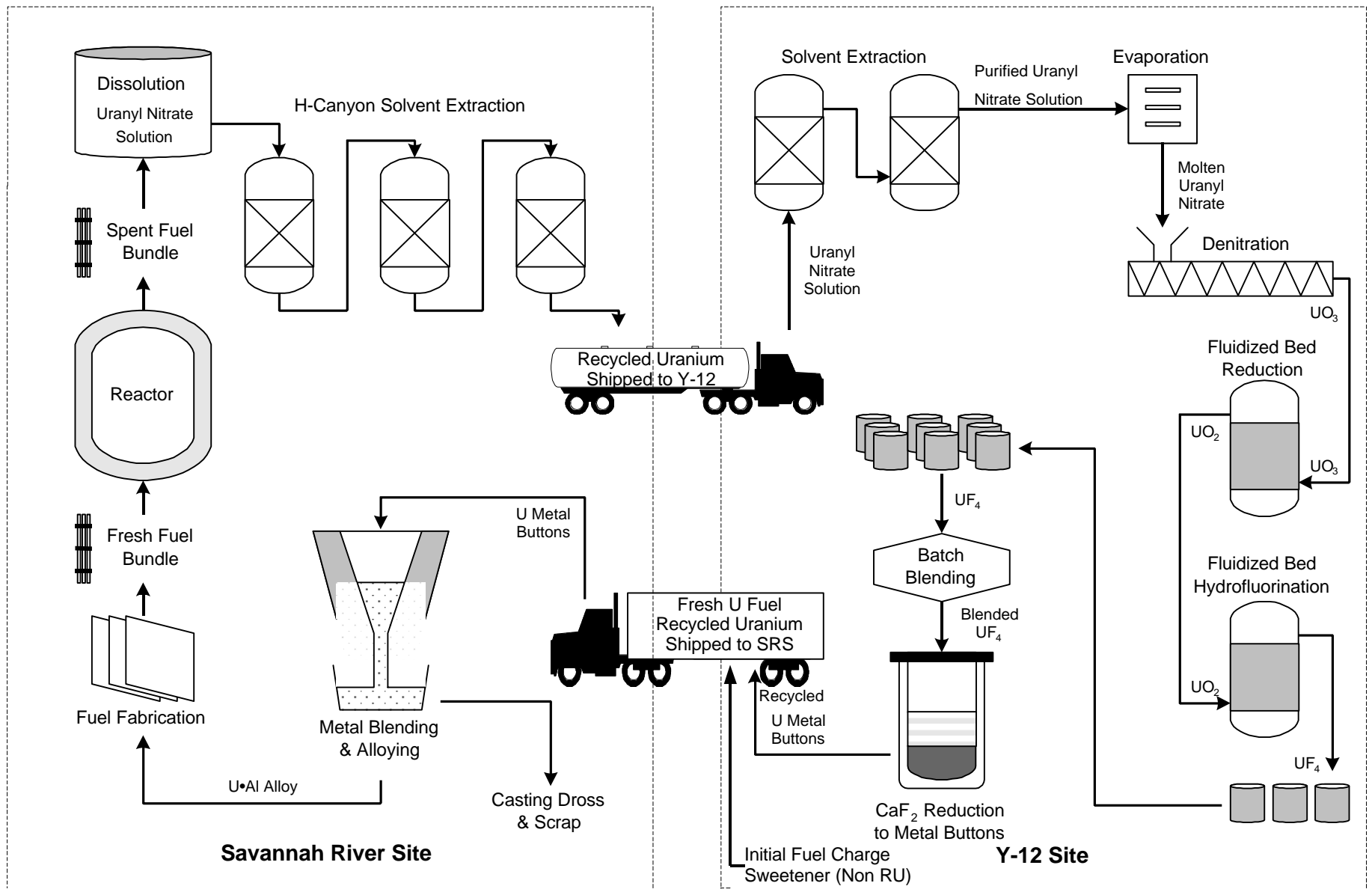


Fig. 3.2-1 Historical Savannah River Fuel Recycle Process

**Table 3.2-1 RU Received at the Y-12 Complex from Source Sites**

Fiscal Year	Hanford		ICPP		Savannah River		Total kg U
	kg U	Assay (%)	kg U	Assay (%)	kg U	Assay (%)	
1953			102				102
1954			231				231
1955			828		2	85	830
1956			744				744
1957	9	0.680	797		3	89	809
1958	1,396	0.650	898		18	82	2,312
1959	82	0.650	3,741		149	83	3,972
1960	15	0.650	769		6,235	85	7,019
1961					2,058	84	2,058
1962			775		2,397	84	3,172
1963					6,446	81	6,446
1964			771		2,978	81	3,749
1965			425		3,552	77	3,977
1966			1,408		3,700	73	5,108
1967					2,502	69	2,502
1968			394		2,109	57	2,503
1969			427		4,090	62	4,517
1970			108		2,060	54	2,168
1971			1,660		3,500	57	5,160
1972			413		4,701	55	5,114
1973			563		5,070	57	5,633
1974					4,581	55	4,581
1975			1,702		5,131	55	6,833
1976			195		4,312	50	4,507
1977			1,333		4,505	45	5,838
1978			526		2,078	47	2,604
1979			535		4,576	48	5,111
1980			(1)		1,489	59	1,488
1981			905		4,911	54	5,816
1982			576		5,719	50	6,295
1983			1,041		6,649	52	7,690
1984			2,868		4,870	57	7,738
1985			1		8,243	52	8,244
1986			960		5,718	56	6,678
1987					4,575	57	4,575
1988					3,095	53	3,095
1989			1		79	66	80
1990					67	66	67
1991							
1992					272	47	272
1993					114	69	114
1994					2,607	64	2,607
1995-1999							
<b>TOTALS*</b>	<b>1,502</b>	<b>0.650<sup>†</sup></b>	<b>25,696</b>	<b>83<sup>†</sup></b>	<b>125,161<sup>‡</sup></b>	<b>60<sup>†</sup></b>	<b>152,359</b>

\* Numbers may not sum because of rounding.

<sup>†</sup> Weighted average

<sup>‡</sup> This number represents only HEU. Another 42.6 MT of LEU was received in 1970 (see text for discussion) and approximately 1 MT of LEU was received over a number of years.

**Table 3.2-2 RU Received at the Y-12 Complex from Secondary Sites**

Fiscal Year	ORGDP		PGDP		TOTAL kg U
	kg U	Assay (%)	kg U	Assay (%)	
1952	1,381	0.650			1,381
1953	2,370	0.641			2,370
1954	137,015	0.673	1,550	0.629	138,565
1955	14,470	0.664	86	0.663	14,556
1956	22,871	0.660	36,440	0.670	59,311
1957	7,588	0.670		0.680	7,588
1958	5,037	0.690	347		5,384
1959	11	0.620			11
1960	2,093	0.640			2,093
<b>TOTALS*</b>	<b>192,836</b>		<b>38,423</b>		<b>231,259</b>

\* Numbers may not sum because of rounding.

RU. A total of 193 MTU was received from ORGDP in the form of oxide and metal. PGDP shipped 38 MT of RU, primarily fluorination tower ash, to the Y-12 Complex. Materials received from Fernald, but not included in this study, were 9,390 MT of DU metal made from gaseous diffusion plant tails for a special project and about 30 MT of LEU.

Some reports indicate that 1.2 MT of UN residue was sent to the Y-12 Complex from West Valley, New York, in 1968. However, this material, recognized as  $^{233}\text{U}$ , was from Consolidated Edison Indian Point-1 Reactor fuel that was recovered by the Nuclear Fuel Services plant at West Valley and sent to Oak Ridge National Laboratory (ORNL) as uranyl nitrate solution for storage. In the 1970s, the ORNL Consolidated Edison Uranium Solidification Program (CEUSP) was initiated to solidify the material for long-term safe storage, and the CEUSP material remains in storage at ORNL today.

HEU material was received at the Y-12 Complex from Rocky Flats for processing as a routine part of the Y-12 Complex mission. Some of this material had surface Pu contamination, but none was identified as RU. Uranium received from Reactive Metals, Inc. (RMI) was low-assay DU that was determined to be below *de minimis* level and so excluded from further consideration.

### 3.3 URANIUM SHIPMENTS

Shipments of RU from the Y-12 Complex are shown in Table 3.3-1. From FY 1961 through FY1989, 120 MTU were sent to SRS. This RU was essentially all HEU metal. In addition, “fresh” HEU (amounting to approximately 70 MTU) was shipped to Savannah River to make up the original fuel charge for the production reactors when they converted to HEU fuel and to be used as sweetener to blend with and enrich the RU to make up the reload fuel elements.

Records indicate that 30 MTU as  $\text{UF}_4$  in the assay range of 0.59 – 0.69% were shipped to PGDP from the Y-12 Complex. It is believed this material was part of the 38 MTU of slightly depleted ash previously identified as RU received from PGDP.

**Table 3.3-1 Y-12 Complex Shipments of RU**

Receiving Site	kg U
ORGDP	192,836
PGDP	29,614
Savannah River	120,384
<b>TOTALS*</b>	<b>342,834</b>

*\* Numbers may not sum because of rounding.*

In addition, data reviewed at the Y-12 Complex indicate a high probability that the 193 MTU received from ORGDP was returned to ORGDP. This material may have been shipped to the Y-12 Complex for temporary storage. Although the data were not conclusive, receipts from ORGDP are assumed to have been returned to ORGDP for the purpose of estimating the Y-12 Complex mass balance receipts and shipments.

Shipments of material from the Y-12 Complex to Fernald, Rocky Flats, RMI, and Hanford were excluded from further consideration in accordance with the DOE Project methodology. The material shipped to Fernald was in a variety of forms, including low-assay DU, LEU, and a small quantity of HEU.<sup>2</sup> The material sent to Rocky Flats was metal apparently made from 0.2% <sup>235</sup>U gaseous diffusion plant tails. The shipments to RMI were low-assay DU billets, and those to Hanford were low-assay DU slugs.

### **3.4 RECYCLED URANIUM WASTE**

Accountability data for uranium, as reported in the documentation reviewed, does not identify losses at a level that can be associated specifically with RU. However, the Project Team was informed by individuals familiar with enriched uranium processing that standards for normal operating losses for once-through processing are approximately 0.5%. During the period of 1953 through 1989, the Y-12 Complex processed approximately 151 MT of highly enriched RU from SRS and ICPP. Solvent extraction raffinate from processing this material was isolated at Building 9206 and transported to the Building 9212 complex. This raffinate was mixed with the raffinate from Building 9212 and processed prior to discarding in the S-3 Ponds. After the mid-1980s, the raffinate was transferred to the West End Treatment Facility. The total RU waste is expected to be less than 100 kg U.

Discussions with individuals who worked at the Y-12 Complex from the 1950s to about 1990 indicated that the plant did not have the capability nor the need to chemically process slightly depleted uranium in the assay range of 0.59 to 0.69% <sup>235</sup>U. It is also known that significant quantities of slightly depleted RU were sent to the Y-12 Complex for storage prior to burial or disposition to other Oak Ridge Operation sites. Therefore, it is assumed that the depleted RU from Hanford, ORGDP, and some from PGDP (a total of approximately 205 MTU) was disposed of in this manner. Since these materials were apparently not processed or handled directly at the Y-12 Complex, they are not considered to be potential sources of personnel or environmental exposure.

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<sup>2</sup> This does not include the slightly enriched RU from Savannah River that was transferred to Fernald immediately upon receipt in the same containers.

### 3.5 RECYCLED URANIUM SCRAP

The primary source of scrap was from the processing of HEU for shipment to Savannah River. Scrap would have been generated in the process of producing metal buttons from the reduction of UF<sub>4</sub> and, in very small amounts, in the fracturing of the buttons to meet the Savannah River specifications. The scrap was recycled to reclaim the uranium. Insignificant losses would have entered the extraction process raffinate stream under normal operation.

### 3.6 INVENTORY AS OF MARCH 31, 1999

As of March 31, 1999, approximately 13 MT of RU in the form of HEU metal buttons and U-Al alloy remained at the Y-12 Complex.

### 3.7 ESTIMATED MASS BALANCE FOR RU

An estimated mass balance for the Y-12 Complex is shown in Table 3.7-1. The mass balance was estimated by comparing the RU received at the plant with a total for the RU shipped, the current inventory, an estimate of RU waste, and depleted RU that was buried or transferred from the plant to other ORO sites for disposition.

**Table 3.7-1 Estimated RU Mass Balance for the Y-12 Complex**

	RU Received (kg U)	RU Shipped (kg U)
Savannah River	125,161	120,384
ICPP	25,696	0
Hanford	1,502	0
ORGDP	192,836	192,836
PGDP	38,423	29,614
<b>TOTAL</b>	<b>383,618</b>	<b>342,834</b>
Total RU Shipped		342,834
RU Inventory (as of 3/31/99)		13,082
Estimated RU Waste		~100
Depleted RU Buried/Disposed		10,311
<b>TOTAL</b>	<b>383,618</b>	<b>366,327</b>
Difference*		~17,300

*\* This difference is due primarily to the inability to precisely distinguish between RU and non-RU shipments.*

The mass balance data reported in this table agree with reconciliation information provided by Hanford and Idaho. The reconciliation also resulted in agreement that material shipped from Fernald was not RU.

The total uranium received from Savannah River was 179 MTU, a difference of 54 MTU when compared with the 125 MTU reported in the table. This discrepancy is explained by receipts from SRS that included 43 MT of LEU received at the Y-12 Complex and which was shipped to Fernald almost immediately. This RU was not removed from the shipping containers nor processed at the Y-12 Complex. Since there is no possibility of Y-12 Complex site environmental exposure, this RU was not added to the various Y-12 Complex report tables representing RU data. In addition, approximately 10 MT of non-RU weapon components sent earlier to Savannah River were returned from SRS and included in receipts, and approximately 1 MTU of miscellaneous LEU was received from SRS.

Total uranium shipped from the Y-12 Complex to SRS was approximately 190 MTU, a difference of 70 MTU when compared with the 120 MTU reported in the table. Savannah River received instructions from DOE that they should report all receipts and shipments involving the Y-12 Complex as RU. The primary item reconciling Y-12 Complex shipments with SRS data is 70 MTU fresh fuel, sweetener, and weapon components determined by the Y-12 Complex to be non-RU. Savannah River also reported small quantities of NU and LEU as RU that were considered by the Y-12 Complex to be non-RU.

A discrepancy of 17.3 MTU between overall receipts and shipments (plus inventory, waste, and buried/otherwise disposed) reflects an inability to precisely distinguish between the RU and non-RU shipments and receipts between the Y-12 Complex and Savannah River and other sites. The only way to distinguish between fresh fuel (non-RU) and sweetener (also non-RU) and RU, using available records, is by enrichment level. The Project Team estimated the enrichment of each shipment and assumed that shipments of  $\leq 90\%$  enrichment were RU. Shipments of  $> 90\%$  enrichment were assumed to be fresh fuel or sweetener, non-RU material. This methodology using enrichment level to distinguish between RU and non-RU results in good estimates of RU flows that are reasonably consistent with Savannah River estimates. Although this is the best available means of distinguishing RU streams, this method does leave a difference of approximately 17.3 MTU between receipts and shipments.

Included in the overall mass balance for RU is highly enriched RU, which is of most concern for worker exposure. An estimated mass balance for just the highly enriched RU is shown in Table 3.7-2. A total of approximately 151 MT of highly enriched RU was received from Savannah River and ICPP, and in return, approximately 120 MT of highly enriched RU was shipped to Savannah River.



**Table 3.7-2 Estimated Mass Balance for Highly Enriched RU**

	<b>RU Received (kg U)</b>	<b>RU Shipped (kg U)</b>
Savannah River	125,161	120,384
ICPP	25,696	0
<b>TOTAL</b>	<b>150,857</b>	<b>120,384</b>
Total RU Shipped		120,384
RU Inventory (as of 3/31/99)		13,082
Estimated RU Waste		~100
<b>TOTAL</b>	<b>150,857</b>	<b>133,566</b>
Difference*		~17,300

*\* This difference is due primarily to the inability to precisely distinguish between RU and non-RU shipments.*